

# Reusing Waste Food-packaging Plastic as Additive Modifier in Asphalt Mixtures

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## Abstract

Packaging waste originated from the food-drinking sector is obtained through the maceration process, which allows the separation of aluminum foil, paper, and plastic. Due to this process, paper and aluminum foil can be reused, while plastic must be disposed. This paper aims to reuse the disposed plastic coming from this process as additive modifier to produce Polymer Modified Asphalt (PMA) mixtures. The research followed chemical and mechanical analyses. The chemical analysis evaluated whether the temperature of production of PMA was enough to melt the waste plastic and studied the chemical composition of the plastic used. Therefore, the melting points of the polymeric components were identified by the Differential Scanning Calorimetry (DSC), Fourier-transform infrared (FT-IR) spectroscopy, and X-Ray Diffraction (XRD) analysis assessed its composition. The mechanical analysis was performed by comparing the PMA mixtures, containing two percentages of reused plastic (5% and 10%), with two mixes prepared with 3.5% Styrene-Butadiene-Styrene (SBS) modified asphalt, using the SuperPave Indirect Tensile Test (IDT) at 10 °C. The DSC highlighted compatibility of the reused material in terms of melting temperatures. XRD and FT-IR results confirmed the waste plastic heterogeneity, presenting mainly Polyethylene (PE), High-Density Polyethylene (HDPE), Low-Density Polyethylene (LDPE), Polypropylene (PP), Polyethylene terephthalate (PET), and Polystyrene (PS). The mechanical analysis showed that 5% plastic PMAs behave similarly to 3.5% linear SBS-modified mixes. However, 10% plastic content decreased PMA workability. In conclusion, it was possible to notice that a 10% increase of plastic in PMAs result in mixes prone to early failure and outside standard parameters.

## Keywords

Waste plastic, Polymer modified asphalt, Workability, Bitumen

## Introduction

The lifecycle of plastic materials is one of the primary challenges in global sustainability. It is also reasonable to assert that the production of plastic materials is essential for the economy, as it is associated with the movement of goods such as food and supplies. The engineering and construction industry offers a potential application field for utilizing these materials after consumption. In fact, engineering innovation is increasingly exploring the use of alternative materials to reduce the depletion of natural resources. The use of plastics without proper recycling plans generates waste disposal problems that many countries currently face. In Europe, for instance, the UK produced approximately 4.9 Mt of plastic waste per year in 2014, which, according to a WWF report, could increase to 6.3 Mt by 2030 [1]. In Italy, 5.9 Mt of polymers were consumed in 2020, predominantly for packaging purposes [2]. Moreover, food packaging plastics account for

more than one third of the production [3], highlighting the need for new disposal strategies to enhance social, economic, and environmental benefits. When considering the asphalt pavement industry, it may present a potential solution to address the issue of disposed waste packaging plastics, thereby reducing costs, and improving environmental benefits. Given the extensive road networks and the need for pavement construction, maintenance, and rehabilitation, a substantial quantity of disposed packaging plastic materials can readily become an easily available resource [4]. However, in this context, it is crucial to ensure that recycled materials exhibit similar behavior to conventional ones [5]. This can be a challenging task due to the heterogeneous composition of food packaging plastics, which plays a key role in terms of both mixing and compacting temperatures. Commonly used polymers for food packaging include LDPE, HDPE, PS, PP, PET, and polyvinyl chloride [6]. These polymers have different melting and/or softening temperatures ranging from 100 °C to 220 °C, which align with the mixing temperature of asphalt materials. In this regard, food packaging plastics can serve as either a modifier of the asphalt binder or a modifier of the asphalt mixture. These two mixing processes are referred to as the wet process and the dry process, respectively, for incorporating food-packaging plastics [6, 7]. In the wet process, the plastic content acts as a modifier of the asphalt binder, similar to the polymer modified bitumen (PMB) procedure. The modified asphalt binder is then used in asphalt plants to produce asphalt mixtures. In contrast, the dry process involves adding the plastic modifier directly to the asphalt plant during the preparation of the asphalt mixture, following the PMA procedure. The main difference between the two lies in the role of plastic. In the wet process, the addition of plastic materials may lead to poor stability of the modified asphalt binder [7]. Nevertheless, plastic modification reduces penetration and ductility, and increases the softening point, viscosity, and high-temperature performance grade of asphalt binders. On the other hand, incorporating plastic content in the mixture acts as part of the aggregate content, binder modifiers, or any combination thereof, reducing susceptibility to permanent deformation, thereby increasing the stiffness and rutting resistance of asphalt mixtures [7]. For this study, the dry process was considered the most suitable and sustainable procedure for reusing waste plastics from food-packaging disposals.

This study aims to evaluate the potential reuse of waste food-packaging plastics, specifically those used for products such as juice, water, and milk, as modifiers for asphalt mixtures using the dry process. To determine the feasibility of this objective, three different asphalt binders were used: one without modification and two with 3.5% SBS modification, consisting of radial and linear SBS polymers, limestone filler, aggregates, and a type of food-packaging plastic. The study initially investigated the composition and temperature-dependent behavior of the food-packaging plastic using FT-IR, XRD, DSC, and Thermogravimetry analysis (TGA). Subsequently, the performance of the PMA was evaluated through the SuperPave IDT test at 10 °C [8-10]. The mechanical analysis was conducted to compare the differences between asphalt mixtures based on PMB and those containing waste food-packaging plastics.

## Materials and Method

### Food-packaging waste plastics

The study proposed aims at re-using plastics coming from multilayered food-packaging carton recycling processes. Basically, cartons are composed of different layers, each one made of different materials, such as different plastics, paper, and aluminum foil. Typically, this kind of waste is recycled through a maceration process, which allows the separation of the different layers, allowing to recover and recycle both aluminum and paper. At the end of this process, the remaining plastic is roughly extruded, thus it is characterised by both polymeric densified grains and plastic fibers, which are typically addressed to incinerators or other recycling chains. This material is mainly characterised by a grain size from 0 to 5 mm, and density from 1.00 to 1.05 g/cm<sup>3</sup>. The materials used were nanoscale.

### Asphalt mixtures and sample preparation

In this study, asphalt mixtures were prepared using two methods: the dry method, which involved obtaining PMA mixtures containing waste plastics, and the traditional method used for reference mixtures. The materials used in the study consisted of three asphalt binders: one neat (N) binder and two 3.5% SBS-modified binders, containing radial (R) and linear (L) SBS polymers, respectively. Additionally, one limestone filler and one type of food-packaging plastic were included. The asphalt binders were characterized by their Performance Grade, which was 58-22 for the neat binder and 64-22 for both SBS-modified binders. It is important to note that the Performance Grade indicates the performance levels of the asphalt binder at service temperatures. The analyzed plastics were used at two different contents: 5% and 10% (by weight of aggregates). Their effects on the performance levels of the mixtures were evaluated by comparing them with the virgin and SBS-modified mixtures, which are referred to as the reference mixtures (RefMix). The RefMix were mechanically mixed to obtain cylindrical specimens with a diameter of 152 mm, following a grading curve (4500 g of virgin aggregates) with a 12.5 Nominal Maximum Aggregate Size, 5.2% asphalt binder content, and a filler-binder ratio of 1.5. The mixing temperatures used were 160 °C for the neat binder and 170 °C for both the L and R binders. Prior to mixing, the binders were placed in a draft oven for at least one hour or until they became fluid. The specimens were then compacted using a gyratory compactor with 126 gyrations to achieve a target air void content of 6% ( $\pm 0.5\%$ ). After compaction, the specimens were allowed to cool down for 12 hours before trimming and sawing to obtain circular-shaped specimens with a thickness of 35 mm. To produce the PMAs, a three-step mixing operation was designed. Firstly, the plastics were added to the pre-heated virgin aggregates. Secondly, the asphalt binders were added and mixed in the bowl. Finally, the filler was incorporated into the mixtures. The PMAs were then placed in a draft oven at 135 °C for 2 hours to simulate the short-term aging process. The material combinations and mixture labels are summarized in table 1.

**Table 1:** Summary of the material combination and mixture labels.

Label	Asphalt binder	Aggregates and filler	Plastic content
LL	3.5% Linear SBS-polymer modified	Limestone	-
NL	Neat	Limestone	-
NP5%	Neat	Limestone	5%
NP10%	Neat	Limestone	10%
RL	3.5% Radial SBS-polymer modified	Limestone	-

### FT-IR spectroscopy

The FT-IR spectroscopy is currently employed to analyze the heterogeneous materials composition through the identifications of their characteristic functional groups. FT-IR spectra, in Total Attenuated Reflectance (ATR) mode directly on powder samples, were collected using the Thermo-Nicolet Nexus spectrometer equipped with Thermo Smart Orbit ATR diamond accessory. Samples were preliminarily subjected to quartering of the material to obtain four representative sub-samples. The spectra were acquired in the 4000 - 400  $\text{cm}^{-1}$  range, with 16 scans and a spectral resolution of 4  $\text{cm}^{-1}$ . For each sample three spectra were recorded.

### XRD

The XRD is a technique used to obtain information about the crystallinity and the crystalline phase of materials. XRD diffractograms were collected by a Thermo ARL X'TRA X-ray diffractometer equipped with a Si-Li detector, at 40 kV and 40 mA, using the Cu-K  $\alpha$  radiation. The samples were scanned in a  $2\theta$  range of 0 - 90°. The peaks identification was performed according to PDF2 database [PDF-2 International Centre for Diffraction Data, 12 Campus Boulevard Newtown Square, PA 19073-3273 USA].

### DSC and TGA

DSC is a thermal analysis technique that studies the influence of temperature variations on the heat capacity ( $C_p$ ) of a material. Changes in the heat capacities of a sample of known mass, when heated or cooled, are plotted as changes in heat flux. This allows to highlight transitions such as melting, glass transitions or phase changes. The TGA is a method to determine the thermal stability of the materials. The variation of mass with temperature is a crucial property of many materials, as they decompose and lose volatile components. This phenomenon can provide useful information about the tested materials. DSC measurements were carried out using a DSC 600 Perkin Elmer instrument in the 10 - 450 °C range, at 10 °C.min<sup>-1</sup> both in nitrogen and air atmosphere. TGA analyses were performed by a Perkin Elmer TGA8000 instrument (mass sample: 1 - 3 mg) at a heating rate of 10 °C min<sup>-1</sup>, on few milligrams of samples, in the temperature range 30 - 550 °C at atmospheric pressure under air flux.

### SuperPave IDT test

To evaluate the PMA mechanical behaviour, the SuperPave IDT test at 10 °C [8-10] was performed. This protocol allows the evaluation of the elastic response, the proneness of materials to accumulate permanent deformation, and the failure resistance and deformation. To assess those, the resilient modulus [8], creep compliance [10], and tensile

strength [9] tests were performed in sequence. In agreement with the PMA Fracture Mechanics Framework [11-13], it is possible to estimate the fracture energy and its components, defined as elastic energy and dissipated creep strain energy, respectively, when the first fracture appears onto the specimen surfaces defining the failure point of the analysed materials. It is worth remarking that for each mixture three replicates were performed.

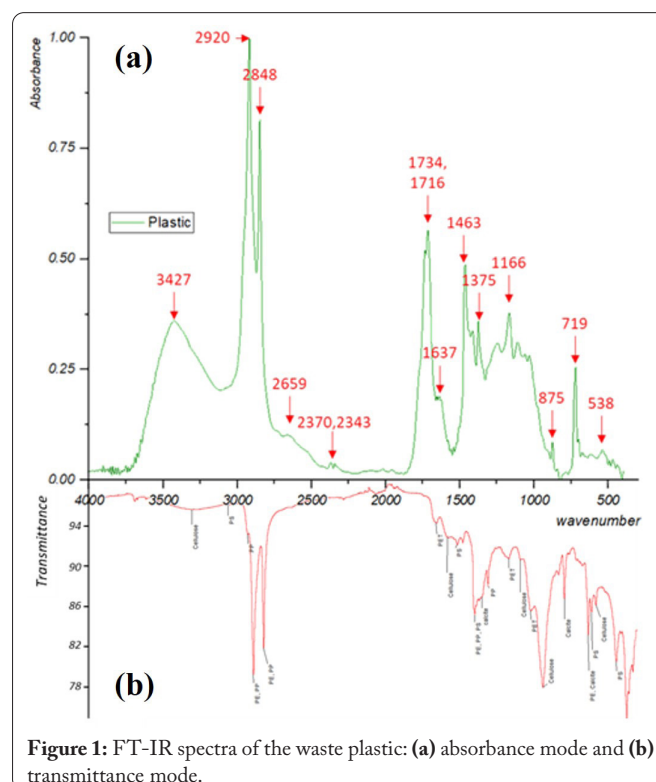
## Results and Discussion

### FT-IR Spectroscopy

Figure 1 shows the FT-IR spectra acquired on the raw samples, in absorbance mode (Figure 1a) and in transmittance mode (Figure 1b). Both spectra are dominated by the absorption bands due to the stretching (at 2920 and 2848  $\text{cm}^{-1}$ ) and bending (in the 1460 - 1300  $\text{cm}^{-1}$  range) vibrations of the  $\text{CH}_2$  and  $\text{CH}_3$  groups of the polymeric chain. By comparison with the FT-IR spectra of different plastics, these features are assigned to LDPE, HDPE, PP. The bands at 1734  $\text{cm}^{-1}$  and at 1637  $\text{cm}^{-1}$  are attributed to the stretching vibrations of the carbonyl groups (C=O) in PET and C=C double bond groups in PS, respectively. In the transmittance spectrum, the functional groups of cellulose and calcium carbonate, belonging to residues of the carton treatment, are highlighted. Centered at about 3430  $\text{cm}^{-1}$ , there is a strong and broad band due to the stretching vibrations of OH groups, involved in intermolecular hydrogen bonds of cellulose [14]. Based on that, it is important to consider how this component might interfere in the blending between binder and plastic when proceeding with the wet method.

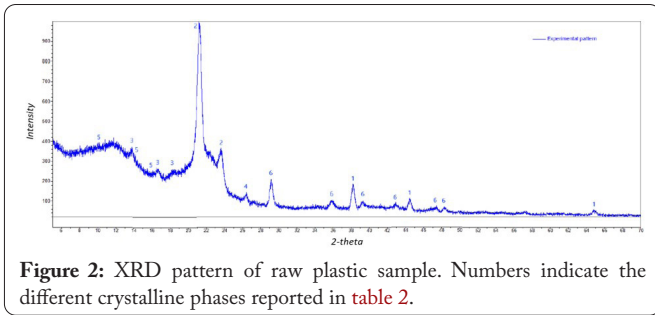
### XRD

Figure 2 shows the diffraction pattern of the waste sample. In the diffractogram, the peaks are marked by numbers



**Figure 1:** FT-IR spectra of the waste plastic: (a) absorbance mode and (b) transmittance mode.



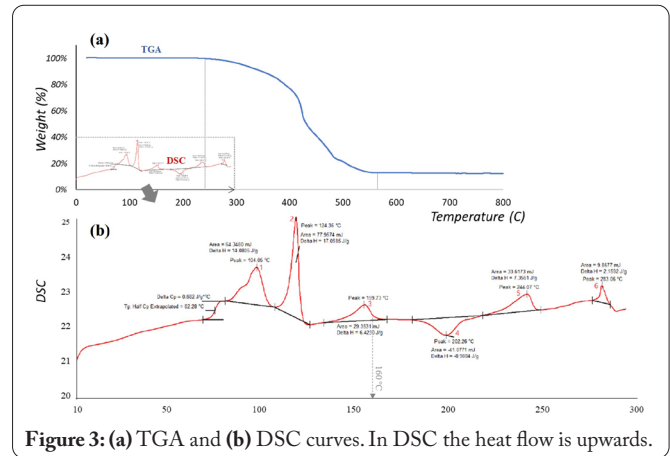


**Figure 2:** XRD pattern of raw plastic sample. Numbers indicate the different crystalline phases reported in table 2.

corresponding to the different crystalline phases, as reported in table 2. The XRD analysis confirms the results obtained with FT-IR measurements: the main polymers identified are PE, PP, and PET. Calcium carbonate is present in the calcite phase, probably used as an inorganic filler in the process of separation of paper from plastic in the raw material. No diffraction peaks attributable to cellulose are identified, indicating that cellulose is present in its amorphous form. Taking into consideration the chemical characterization it is relevant to note that the material has still residue of cellulose and aluminum. These components might interfere with the asphalt binder.

**DSC and TGA**

Figure 3 presents the TGA and DSC profile of the waste samples. The TGA curve (Figure 3a) presents different stages of mass loss as a function of the heating temperature, giving an indication of the inhomogeneity of the material. This result is expected, considering that the waste plastic used in this research is composed of different types of polymers [15, 16]. Thermal decomposition occurs between 200 and 550 °C. The first process, between 200 - 350 °C, is mainly due to the degradation of cellulose components. At about 380 °C the thermal degradation of the plastic components begins up to about 520 °C showing a wide range of degradation temperatures. PS degrades at the lowest temperature (380 - 400 °C) among the plastic components followed by PET (420 °C) and PP (450 °C) while HDPE degrades at the highest temperature range (480 - 550°C) [17]. The DSC curve (Figure 3b) shows four endothermic peaks associated to the solid-liquid phase transition.



**Figure 3:** (a) TGA and (b) DSC curves. In DSC the heat flow is upwards.

LDPE, HDPE, and PP exhibit low melting points with peak temperatures at about 104, 124, and 159 °C, respectively [18]. The other 2 endothermic processes, at temperatures over 200 °C, are related to the melting point of PET, at around 244 °C and PS at around 283 °C.

When it comes to materials that are used for asphalt mixing, it is important to consider the temperatures that the mixtures need to achieve during construction. It must be considered that these components hardly melt during the process of blending with the binder. In this case, even when applied by the wet process, part of the waste plastic might still stay dispersed inside the mixtures working as aggregate.

**Mechanical analysis**

Table 3 presents the summary of the results obtained performing the SuperPave IDT test at 10 °C. Considering the resilient modulus, the highest value (21.70 GPa) was shown by NL. Considering that the resilient modulus depends by aggregates skeleton, the outcome obtained for NL is likely linked to an over compacted condition. In fact, the others present a modulus between 9 and 13 GPa. It is important to highlight that the NP5% are characterised by the higher modulus than NP10%, highlighting that 5% of plastic allows to obtain an aggregate skeleton like the virgin mixture containing PMBs. Conversely, considering the creep

**Table 2:** Crystalline phases, diffraction angles, and reflection planes.

Crystalline phases	2θ diffraction angle	Reflection plane	Database code
Aluminum	38.4; 44.6; 65.1	[111]; [200]; [220]	JCPDS: 01-088-3657
PE	21.5, 23.8; 36.1	[110]; [200] [020]	JCPDS: 00-040-1995
PP	14.0; 16.8; 18.4	[110]; [040]; [130]	JCPDS: 00-050-2397
PET	26.1	[100]	JCPDS: 00-049-2301
Calcite	29.3; 36.0; 39.4; 43.2; 47.4; 48.5	[104]; [110]; [113]; [202]; [018]; [116]	AMCSD: 0000985

**Table 3:** Summary of mechanical analysis.

Materials	Resilient modulus (GPa)	Creep compliance (1/GPa)	m-value	Tensile strength (MPa)	Failure strain (µstrain)	Fracture energy (kJ/m³)	Elastic energy (kJ/m³)	Dissipated creep strain energy (kJ/m³)
LL	10.50	3.07	0.41	3.36	0.87	1.84	0.54	1.30
NL	21.68	1.57	0.52	4.64	0.66	1.85	0.50	1.35
NP5%	12.98	1.05	0.31	3.06	0.75	1.44	0.36	1.08
NP10%	9.51	0.31	0.19	2.68	1.33	2.26	0.40	1.85
RL	12.38	1.39	0.34	3.23	1.11	2.22	0.44	1.78

compliance test, the addition of plastic highly influences the accumulation of the permanent deformation. In fact, NP5% and LL were characterised by quite the same creep compliance and m-value, meaning that the added plastics reduce the proneness to accumulate permanent deformation similarly to the PMB containing radial SBS. Particularly, NP10% exhibits the lowest creep compliance and m-value highlighting the elastic action of the added plastics. In terms of tensile strength, the highest value (4.64 MPa) characterised NL. Nevertheless, the low failure strain indicates a quite brittle failure. On the other hand, the lowest tensile strength result was observed for NP10%, which also exhibited the highest deformation at failure point, indicating a low level of mixture compaction. Considering the energetic parameters, it is possible to highlight the low fracture energy of NP5%. This is likely linked to limited deformation at failure point.

## Conclusions

This study evaluates the re-use of food-packaging waste plastics from the carton recycling process. The analysis includes chemical and mechanical assessments. The chemical analysis examines the plastic's characteristics and thermal properties for producing PMA mixtures. The mechanical analysis involves Superpave IDT testing at 10° to evaluate the mechanical and cracking properties of the mixtures. The key findings are as follows:

- The food-packaging waste plastics are heterogeneous, consisting of various polymers, residual cellulose, calcium carbonate, and aluminium.
- Despite the heterogeneous composition, the waste plastics have compatible melting points for mixing and compacting PMA mixtures.
- Mixtures with food-packaging plastics demonstrate similar performance to those containing 3.5% SBS modified asphalt binder. This suggests that the mixing procedure and materials used are suitable for preparing PMAs.
- However, as the plastic content increases to 10%, PMAs fail to meet the required standard parameters in terms of air void content, resulting in premature failure of the mixtures.

## Further Improvements

Further improvements will be focused on volumetric analyses of PMAs containing plastics in order to study their workability during mixing and compacting operations. Moreover, a comparative analysis will be focused on the re-use of untreated and pelletised food-packaging waste plastics using the same approach described and discussed above.

## Acknowledgments

None.

## Conflict of Interest

None.

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